ATOMISTIC SCALE STUDY OF LOCAL HEATING INTO SILICON:NANOINDENTATION USING MOLECULAR DYNAMICS SIMULATION

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ABSTRACT.*This paper presentsthe atomistic scale study of the effects of local heating during nanoindentation into Silicon (Si). Nanoindentation techniques and experiments with fundamental principles of MD simulations were at first introduced.The indentation processesof indentation depths an order of magnitude much smaller than experimental values and over indentation times several orders of magnitude shorter. The effects of local heating were studied using Force-Depths (F-D) curves and snapshots. The influences of the indentation speeds and temperature on the deformative behaviour of Silicon (Si) were also discussed. The force-depth curves signified the temperature influence on silicon during retraction for thermalized speeds, implying that the topology of the silicon surface did not change after tip retraction. Snapshots of the simulations into Si showed that slower indentation speeds would exhibit a decrease in the number of heated atoms.*

KEYWORDS. Nanoindentation, indentation depths, indentation speeds, thermalized,simulations.

INTRODUCTION

Nanoindentation is a technique to determine the mechanical behaviour of materials on the nanometre scale. Numerous experiments using nanoindentation have identified a remarkable phenomenon, namely, a systematic study using DSI (Depth Sensing Indentation systems) or nanoindenters on unidirectionally patterned aluminium lines on Silicon (Si) substrate had been conducted to understand the microplasticity at the incipient stage of nanoindentation, as well as the effects of length scale on the elastic deformation of materials(Choi Yoonjoon and Suresh, 2003). Studies by Landman*etal*. (1989)and Shimizu *et al.*(1998) had reported MD simulations of nanofriction for Si-Si and diamond-Copper (Cu) interfaces which observe the stick-slip phenomenon, that is, the transition from static to dynamic motion.

MATERIAL AND METHOD

Using modern nanoindentationequipments to determine the mechanical properties of material is relatively easy; however it is quite difficult to understand the mechanics and nature of plastic deformation of materials. Hence, computer simulations based on approaches such as molecular dynamics, offer an ideal and popular approach to investigating numerous questions that arise and cannot be fully explained from experimental nanoindentation measurements, specifically the shape of the Force-Depth (F-D) curves of the simulations that can be related to specific atomic scale events during indentation. In nanoindentationexperiments, the

procedure involves pressing a nanosized tip at a constant speed *v* from a given height to a given depth into the surface of a material. This process is called loading. It is then subsequently retracted to its original position following the same path as for the loading. This retraction process is the called unloading. The measurements involve very small penetration forces, which will result in very small indention depths of nanometre scale in the material investigated.

MD SIMULATION METHOD

Equations of Motion for Atomic Systems

Considering a system containing N atoms of which the potential energy would be the summation of individual atoms, pairs, triplets,etc.

$$\mathrm{V}=\sum_{i} v_i(r_i)+\sum_{i}\sum_{j>i} v_2(r_i,r_j)+\sum_{i}\sum_{j>i}\sum_{k>j>i} v_3(r_i,r_j,r_k) \qquad (1)$$

The equation of motion is thus, a Lagrangian function and can be defined in terms of kinetic energies, *K*,and potential energies, *V*,such as $L = K - V$. It is thus considered to be a function of the generalized coordinates, q_k and their time derivatives, q_k'. The Hamiltonian form of the equation of motion can therefore defined by equation (2):

$$\mathrm{H}(p,q)=\sum_{k} q_k' p_k - \mathrm{L}(q,q_k') \quad (2)$$

In atomic systems, with Cartesian coordinates, r_i and the usual definitions of K and V, with $F_i = \nabla_{r_i} \mathrm{L} = -\nabla_{r_i} \mathrm{V}$ is the force on that atom at location r_i . Then the equation of motion for the *i*th atom wouldthusbecome:

$$m_i a_i(t) = F_i \qquad (3)$$

with, $m_i$as the mass of atom, i, and $a_i(t)$, its acceleration.

In this study using MD simulation, the Born-Oppenheimer approximation was used. Realistic many-body interatomic potentials were used to describe the system and the forces on the tip, calculated by summing the total forces acting on the individual atoms. Examples of potentials used in these simulations were the Brenner potential (Brenner, 1990; 1998) for the silicon-silicon (Si-Si) interaction, the Tersoff potential for the C-Si interaction (Tersoff, 1988;1989). The diamond-tip indenter which had a carbon-carbon(C-C) interaction was based on the Brenner potential (Brenner, 1990;1998). Initially adopted by Verlet(Verlet, 1967), the position of every particle i at time t was read as follows:

$$r_i(t+\delta t) = 2r_i(t) - r_i(t-\delta t) + a_i(t)\delta t^2 \qquad (4)$$

Here, $a_i(t)$ was calculated using equation (3). The velocities could determine the kinetic energies of the atoms. Swope *et al.* (1982) had proposed the 'velocity' Verlet algorithm which did not need the storage of the position at time ($t-\delta t$), in which the new position and velocity of atoms could be computed as follows:

$$r_i(t+\delta t) = r_i(t) + v_i(t)\delta t + 1/2 a_i(t)\delta t^2 , \qquad (5)$$

$$v_i(t+\delta t) = v_i(t) + 1/2[a_i(t) + a_i(t+\delta t)]\delta t . \qquad (6)$$

Since the total energy was not exactly conserved for a few MD time steps, it oscillated very closely to a mean value and therefore it could be said that on average the energy was

conserved over time scales of a few hundreds of time steps. This algorithm formed the time integration of the atomic coordinates in the MD simulations of this study.

Boundary Conditions

For crystalline materials, boundary conditions were employed to maintain as accurately as possible the realistic physical behaviour of the simulated material. There were typically four types of boundary conditions which could be imposed on the atoms in the system and were specific to the kind of phenomenon under study. Free boundary conditions were usually applied when there were no restrictions on the dynamics of the atoms while fixed or rigid boundary conditions could be applied to edge atoms to constrain the vertical or horizontal motion of the atoms. Edge effects could be reduced by employing periodic boundary conditions (PBCs) that allowed edge atoms to interact with those on the opposite side of the lattice.

A standard Nosé–Hoover thermostat (Hoover, 1986) was used to control or damp the temperature of the system by simulating effectively the dissipation of energy through the bulk. Periodic Boundary Conditions (PBCs) were applied to all the side atoms of the substrate while all the other atoms were allowed to move with the MD algorithm.The silicon substrates were indented initially, at first without temperature, that was, damped and later, the materials were thermalized with temperature added to the system by a heat bath until 300K. Then, the heat bath was removed and thermalization only occurs at the sides of the substrates during indentation. This is done so that the heat generated during indentation when the atoms were damped and thermalized could be distinguished. The damping model was not based on any concept of temperature control and for simplicity the Lindhard-Scharff inelastic loss model (Lindhard*et al.*, 1963) was used.

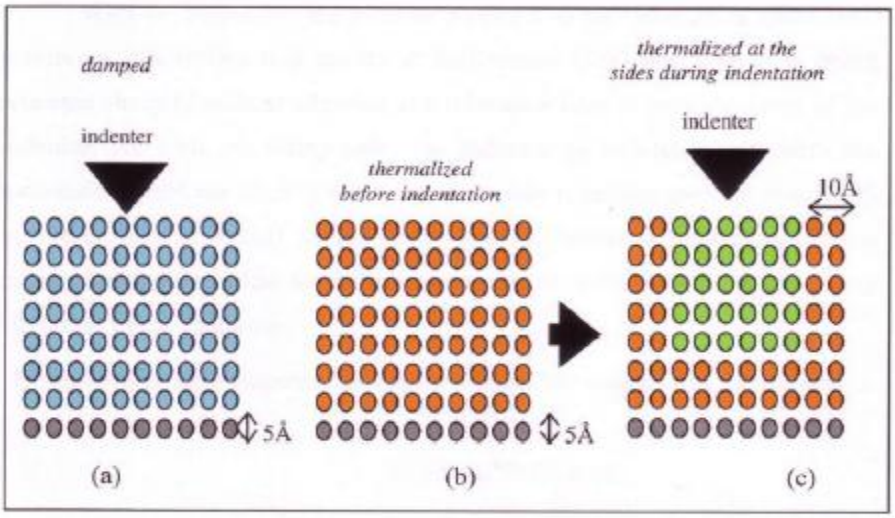

Figure1. A schematic diagram of substrate conditions utilized in the nanoindentation simulations. The grey shaded atoms denote 5Å of fixed atoms, (a) blue-damped atoms, (b) red-thermalized atoms at 300K before indentation, and (c) green-thermalized atoms 10Å above the fixed atoms and round the sides of lattice only during indentation.

Thermalized Atomic Motion of Equations

The temperature of the systems of many physical conditions might be held fixed rather than conserved the total energy. This could be achieved by maintaining the system in contact with a thermal bath. The atoms then followed a random motion similar to that described by the Brownian movement, that was, small particles of colloidal size immersed in a fluid colliding with the microscopic particles of the fluid. The equation of motion for a particle i immersed

in a fluid was given by the Langevin equation (Doob, 1942):

$$ma_i(t) = F_i - \beta r_i'(t) + \Gamma_i(t) \qquad (7)$$

where three different terms were clearly distinguished, namely: i) the first term was related to the interatomic interactions, ii) the second term representing a dynamical friction due to the viscosity of the surrounding fluid, and iii) one stochastic part, being the characteristics of the Brownian motion with a zero mean, $<\Gamma_i(t)>=0$.

For a system in thermal contact with a heat bath, the direction and velocity of the particles were continuously being modified by the exchange of phonons between the system and the heat bath. Thus phonons displayed the Brownian motion as that of the microscopic particles immersed in a fluid. Ermack(1976) had developed an algorithm to conduct 'Brownian dynamics' simulation by which the position and velocity at a time t could be advanced as follows:

$$r_i(t+\delta t) = r(t) + c_1 v_i(t)\delta t + c_2 a_i(t)\delta t^2 + \delta r_i^G \qquad (8)$$

$$v_i = c_0 v_i(t) + (c_1 - c_2)a_i(t)\delta t + c_2 a_i(t+\delta t)\delta t + \delta v_i{}^G \qquad (9)$$

wherethe values of the coefficients were :

$$c_0 = e^{-\alpha\delta t} \qquad (10)$$

$$c_1 = (\alpha\delta t)^{-1}(1-c_0) \qquad (11)$$

$$c_2 = (\alpha\delta t)^{-1}(1-c_1) \qquad (12)$$

and δr_i^G and $\delta v_i{}^G$ were random variables sampled from a bivariate Gaussian distribution (Chandrasekar, 1943). By varying the sequence of random numbers δr_i^G and $\delta v_i{}^G$, different trajectories could be simulated. However, these simulations would all lead to the same thermodynamics averages after a sufficiently long time.

Nanoindentation of the Si substrates as a function of the indentation speed, *v* was run using a Ramp-coded velocity profile which allowed the slowing down or regaining speed of the indenter just before and after maximum indentation, that was, 10% on either side of a half period (T/2), during loading and unloading. This allowed the indenter to indent or retract without any strong vibrations of the cantilever. The simulations ran from a few hours to a few weeks, for slow indentation speed, depending on the amount of computer power available in each workstation. A schematic diagram of the Ramp-MD code velocity profile was shown in Figure 2 that follows.

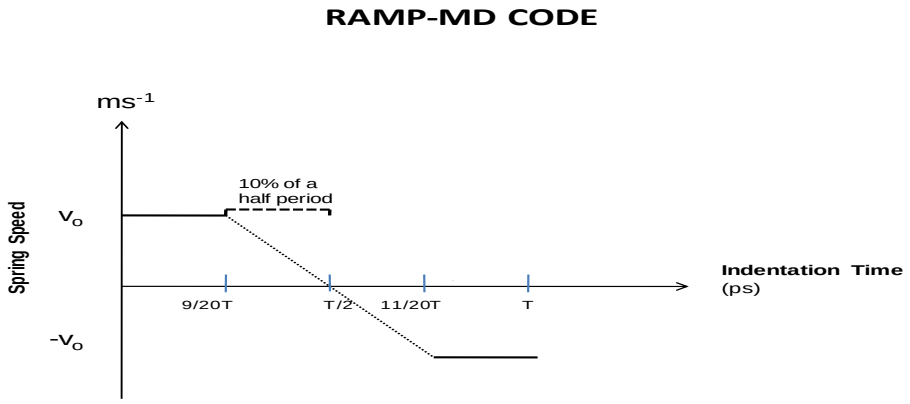

Figure 2: Spring Speed versus Indentation Time for Ramp-coded MD simulations

RESULTS AND DISCUSSIONS

MD Simulation

The workspace parameters employed for the Si substrates were depicted in Table 1. The simulation snapshots pictures presented were produced using VTK (http://www.kitware.com/vtk).In the snapshots, atoms were presented as spheres and coloured according to their local heating, that is, temperature of the atoms during indentation.

Table 1: Workspace and simulation parameters for silicon substrate

Substrate material	Silicon
Substrate crystal structure	Diamond-like
Workspace dimensions	65Å x 63Å x 65Å(001) surface 13824 atoms (1647 tip atoms)
Indenter directions	[001]
Indenter speeds	5m/s and 50m/s
Substrate temperature	Damped (zero K); Thermalized (300 K)
MD time step	1.00 fs

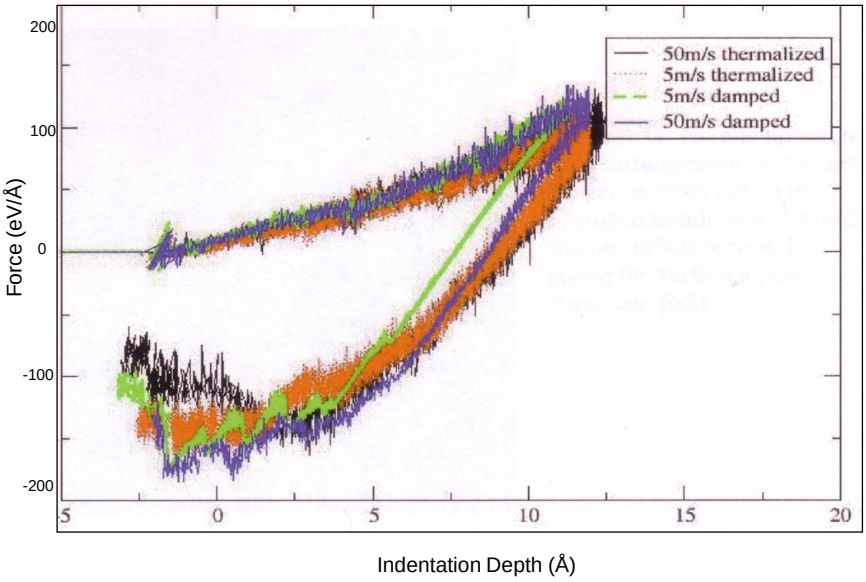

Figure 3. Force-Depth Curve for Silicon at 5 and 50m/s damped and thermalized speeds

The force-depth curves for silicon using the Ramp-code, at two indentation speeds, 50 and 5m/s respectively, damped and thermalized, were shown above in Figure 3. The maximum force for both sets of speeds is about 125eV/Å; with damped speeds at approximately 11Å of depth and thermalized speeds at approximately 12Å of depth. The damped speeds depicted a steeper slope during loading and unloading. The sudden retraction of the indenter during unloading might cause the silicon surface to end in an amorphous structure, thus a steeper slope as shown in the graph. During unloading at both thermalized speeds, less force was required. This was due to fact that higher energy states between the tip atoms and the surrounding locally heated atoms being able to break easily from their binding forces. With thermalized speeds, however, the curves showed an approximately exact height and gradient at both loading and unloading stages. The surrounding atoms, energized by the exchanged of energy from the thermalized atoms, were able to move around their positions due to the extra vibrations, hence allowing the indenter tip to penetrate deeper into the material. There was a recovery to a nearly flat surface during unloading due to the thermal vibrations of these atoms at 300 K. This implied that the topology of the silicon surface did not change after tip retraction. This phenomenon had also been reported that MD simulations with a hydrogen terminated dimer reconstructed silicon surfaces (Sanz-Navarro *et al.*, 2004) had shown a remarkable influence of the atomic thermal vibrations in the recovery of the indented region during unloading. A snapshot presentation of the hydrogen terminated (100)-(2x1) silicon surface at peak load was shown in the Figure 4 below.

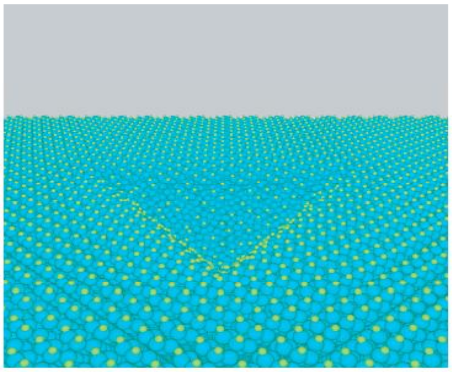

Figure 4.No pile up and no real rearrangement of the surface dimers were observed on the hydrogen terminated (100)-(2x1) silicon surface at peak load giving the surface a pure distortion (Sanz-Navarro*et al.*, 2004).

Figure 5(a) and Figure 5(b) below shows the snapshots of simulations for 50 and 5m/s damped speeds for silicon at maximum indentation times in the temperature range of 0 to 150 K. It is noticed that there is a presence of some heated atomsat 50m/s but virtually none at 5m/s, implying that the thermostat has successfully dissipated the heat generated locally during indentation at 5m/s.

The following Figure 6(a) and Figure 6(b) also shows the snapshots of simulations for 50 and 5m/s thermalized speeds at maximum indentation times in the temperature range of 0 to 750 K. The system is annealed to 300 K, hence the presence of heated atoms at 50m/s being obviously seen to be more than at 5m/s thermalized, and when compared with the damped speeds. Again, the thermostat has successfully dissipated the heat generated during indentaton at the slower thermalized speed of 5m/s.

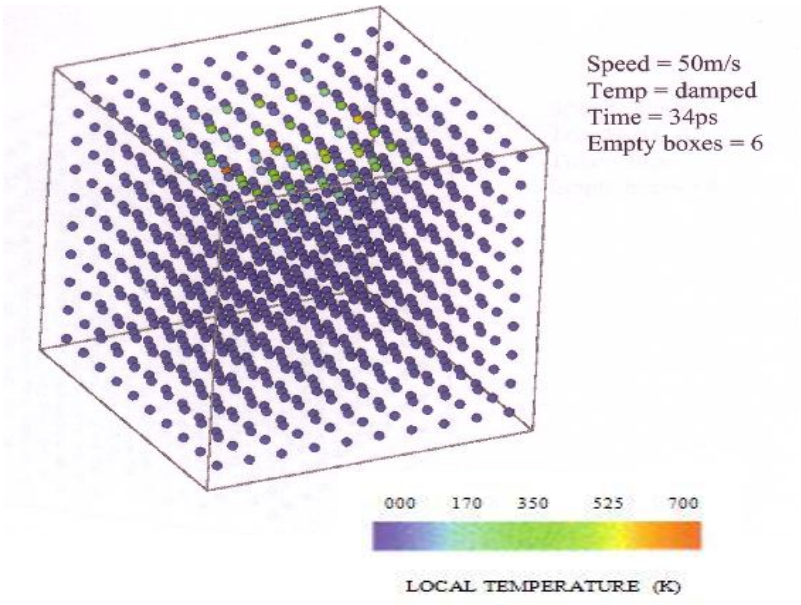

Figure 5(a): Snapshot for damped speed of 50 m/s for silicon at maximum indentation time

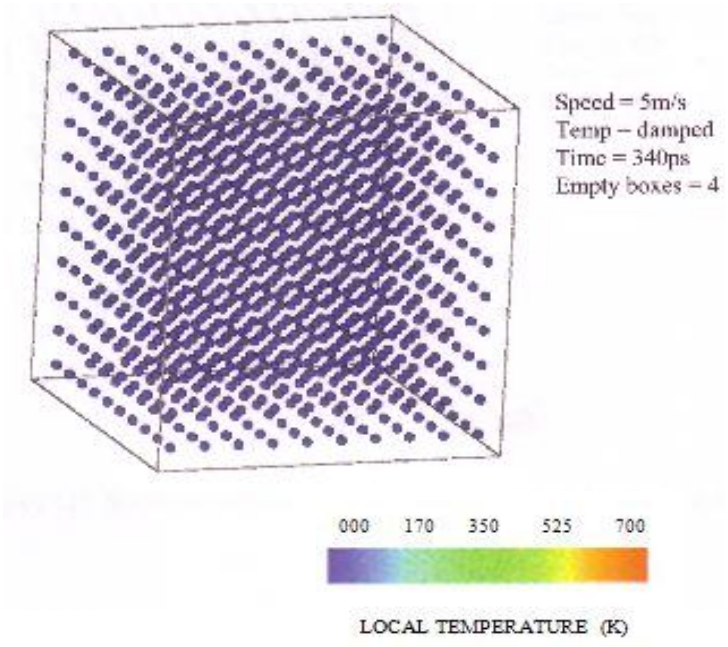

Figure 5(b): Snapshot for damped speed of 5 m/s for silicon at maximum indentation time

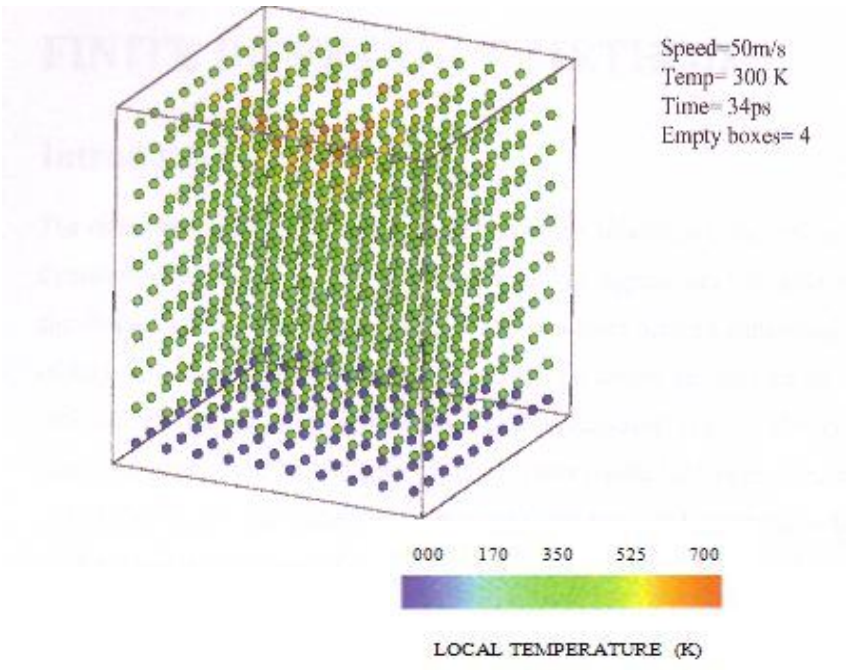

Figure 6(a): Snapshot for thermalized speed of 50 m/s for silicon at maximum indentation time

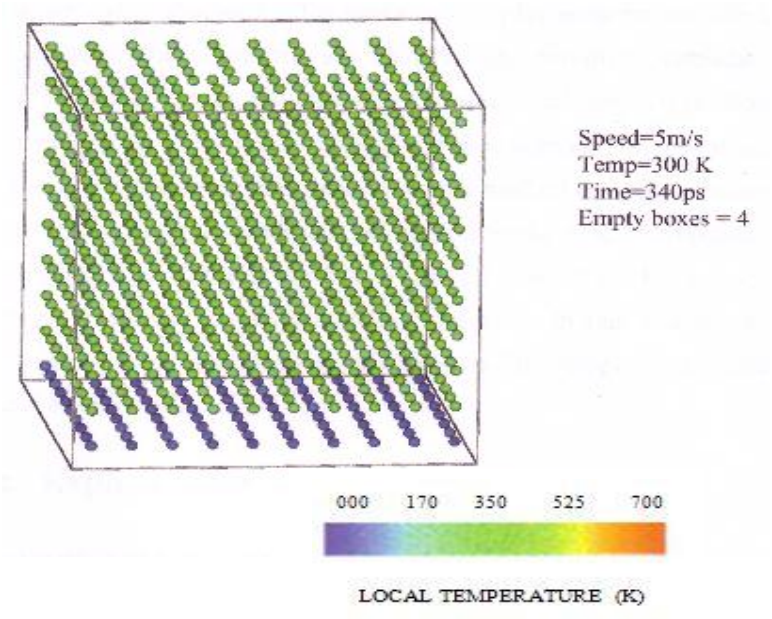

Figure 6(b): Snapshots for thermalized speed of 5 m/s for silicon at maximum indentation time

CONCLUSION

For silicon, at both thermalized and damped speeds of 50 and 5 m/s, slightly different characteristics were exhibited, in accordance to the laws of physics. At thermalized speeds, the force-depth curves signified the temperature influence on its transitive behaviour during retraction. The silicon surface was able to recover to a nearly flat surface due to the vibrations of the thermalized atoms. The indenter was able to penetrate deeper into the Si substrates due to fact that there was more local kinetic energies below the indenter and hence, in the substrates at thermalized speeds. However, at damped speeds, sudden retraction of the indenter resulted in the insufficient time for the atoms to exhibit similar effects as with the thermalized speeds, hence depicting a steeper slope during unloading. Snapshots of the MD simulations of Si also showed that the number of heated atoms would decrease with slower indentation speeds for both damped and thermalized. For future work, it was recommended that slower indentation speeds such as 5m/s, would best be used in MD simulations of nanoindentation on materials of composite structures too. Further researchonnanoindentation into bio-nanomaterials of different molecular structures would certainly gave an insight of the indentation speeds, indentation times, behaviour of the heated atoms on temperature, and others, besides the substrate surface nucleation.

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